

APPENDIX 3: CTV II GEOCHEMICAL MODELING

Memorandum

To: Travis Hurst, CRC

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From: Chris Wolf, P.G. and Beth Salvas, P.G.

Subject: CTV II Geochemical Modeling

1. Introduction

For a proposed carbon sequestration project CTV II, CRC has requested that Daniel B. Stephens & Associates, Inc. (DBS&A) perform geochemical modeling to help understand chemical reactions during carbon dioxide (CO₂) storage. Information used to perform the modeling described in this memorandum was provided by CRC.

Geochemical modeling was conducted to evaluate the compatibility of the injectate with groundwater and rocks comprising the Injection Zone and Confining Zone. The intent of the modeling is to identify the major potential reactions that may affect injection or containment (U.S. EPA, 2013).

Geochemical modeling using the PHREEQC (pH-REdox-Equilibrium) software was used to calculate the behavior of minerals and changes in aqueous chemistry and mineralogy based on chemical equilibrium conditions (Parkhurst and Appelo, 2013).

This technical memorandum describes the site conditions and modeling results for CTV II (Tables 1 through 7).

Based on the geochemical equilibrium modeling, the injection of carbon dioxide at the CTV II site does not cause significant reactions that will affect the injection or containment of the gas.

2. Geochemistry for CTV II Storage Project

While rocks are buried in the earth's crust, chemical reactions between the rocks and groundwater are termed diagenesis, which involves the dissolution of minerals into groundwater and precipitation of minerals from solution. Reactions are driven by fluid movement, temperature, and pressure changes due to burial depth and compaction. Over time, minerals

and cements may dissolve and form new minerals. Important reactions that typically occur in clastic sedimentary rocks include the following:

- Precipitation and dissolution of cements and authigenic minerals consisting of various minerals including quartz, clays, potassium feldspar (K-feldspar), plagioclase feldspar, siderite, gypsum, and pyrite
- Dissolution of feldspars, quartz, lithic fragments
- Formation of feldspar and quartz overgrowths
- Precipitation of illite, kaolinite and other clays

2.1 Injection Zone Fluid Geochemistry

Data from water samples collected in the Injection Zone in CTV II (Table 1) were used for the geochemical modeling because they included a complete suite of major ions and pH. With a total dissolved solids (TDS) concentration of 15,595 parts per million (ppm), the groundwater is considered brackish.

The net charge of a water sample may be calculated using the results for the cation and anion data. Because water has a net neutral charge, the sum of the cation and anion charges should be zero. Variations due to sampling and analyses often cause the calculated value to vary, and a value within 5 percent of neutral is considered a “good” balance. The charge balance for the water sample was calculated in PHREEQC at 5.3 percent. The charge balance was subsequently corrected in the models by allowing PHREEQC to alter the sulfate (SO_4) concentration to maintain the charge balance.

2.2 Injection Zone and Confining Zone Mineralogy

Mineralogy was evaluated using x-ray diffraction (XRD) to determine the bulk and clay mineralogy of core samples.

At CTV II, mineralogy of the Injection Zone is dominated by quartz and feldspars, with about 10 percent clay mineral content (Table 2). One sample in the injection zone is dominated by 58 percent clay minerals. The Upper Confining Zone is dominated by 40 to 60 percent clay minerals, with lesser amounts of quartz and feldspar.

2.3 Injectate Chemistry

For the geochemical modeling, two scenarios of different chemical compositions for the carbon dioxide injectate were developed (Table 3). The compositions were normalized to 100 percent for use as model input. For Scenario 2, the ethane component was excluded from the geochemical analysis because ethane gas is not in the model database. The chemistry for Scenario 1 and Scenario 2 was modeled at CTV II.

3. Equilibrium Geochemical Modeling

When modeling groundwater geochemistry, the water chemistry, gas chemistry, and mineralogy are used to constrain the model because mineral solubility controls the concentrations of a mineral's elemental components in groundwater (Appelo and Postma, 2005). Mineral dissolution-precipitation reactions directly impact the aqueous chemistry. In general, as minerals dissolve the elemental concentrations in groundwater increase, and when minerals precipitate the elemental concentrations in groundwater decrease. Chemical equilibrium indicates that congruent reactions will appear balanced between reactants and products, with no apparent change in the chemical system.

The PHREEQC model was used to evaluate potential changes to mineralogy and aqueous composition in the subsurface due to CO₂ injection. The mineral, gas, and aqueous phases were assumed to be in chemical equilibrium.

Based on the available injectate gas compositions, the ideal gas law and Raoult's Law were used to calculate the gas composition in moles. The initial and final pressures of 81.7 and 308.7 atmospheres (atm) at CTV II were used to calculate the partial pressures of the injectate components.

A reservoir temperature of 103°C was used for CTV II.

3.1 Geochemical Database

For reactions involving water and minerals, the equilibrium relationship between products and reactant activities (concentrations) can be calculated using known values for parameters like Gibb's energy found in thermodynamic databases (Zhu and Anderson, 2002). Thermodynamic values for these calculations are compiled in databases from several entities, including the U.S. Geological Survey (USGS) and Lawrence Livermore National Laboratory. A database developed at the Lawrence Livermore National Laboratory (LLNL.dat) was used for this

evaluation. The LLNL.dat database includes a temperature range for the thermodynamic data provided from 0 to 300°C. This database is appropriate for the groundwater concentrations, pressure, and temperature used in the modeled scenarios.

When modeling saline waters, the Pitzer database (Parkhurst and Appelo, 2013) is often used, but it has thermodynamic data for a limited number of minerals including calcite, dolomite, gypsum, and quartz. The Injection Zone and Upper Confining Zone include minerals that are not included in the Pitzer database, so the LLNL.dat database was used because it also includes smectite, illite, pyrite, and the minerals listed in Table 2.

For the injection gases, methane is included in the database as a gas and aqueous phase, but ethane is not included as a gas phase. The ethane gas portion of the injection chemistry was not modeled. The mass fractions were normalized excluding the ethane.

3.2 Saturation Indices

Saturation indices (SIs) were calculated that represent whether a particular mineral (e.g., calcite or gypsum) is in chemical equilibrium with the groundwater. SI calculations are used to predict if a mineral is likely to precipitate or dissolve in the groundwater, and if these reactions change the concentrations of dissolved elements.

Chemical equilibrium was assumed for the reactions in the model. Equilibrium modeling sets the saturation indices to a zero (0) value for a given mineral. Minerals used in the modeling scenarios are based on those detected using XRD and their relative abundances. The assumption of chemical equilibrium allows dissolution and precipitation reactions to be quantified in the model.

The formula for calculating saturation indices (SI) is as follows:

$$SI = \frac{IAP}{K_{sp}} \quad (1)$$

where SI = saturation index

IAP = ion activity product

K_{sp} = solubility product

Using gypsum as an example (Clark, 2015), the ion activity product of gypsum (IAP_{gypsum}) is the product of the activity (a , activity is approximately equal to concentration in dilute solutions) of calcium (Ca) and sulfate (SO_4):

$$IAP = a_{Ca^{2+}} \times a_{SO_4^{2-}} \quad (2)$$

The solubility product, K_{sp} , is an indication of the relative solubility of a mineral in water. A value less than zero (<0) indicates that the mineral will dissolve and contribute ions to solution and may result in a relatively high activity or concentration. A value greater than zero (>0) indicates that the mineral has a low solubility, may precipitate from solution, and will not contribute many ions to the solution. For the mineral gypsum, the K_{sp} based on the dissociation reaction of gypsum in water is calculated as follows:

$$CaSO_4 \cdot 2H_2O \leftrightarrow Ca^{2+} + SO_4^{2-} + 2H_2O$$

$$K_{sp} = \frac{a_{Ca^{2+}} + a_{SO_4^{2-}} + a_{H_2O}}{a_{gypsum}} = 10^{-4.60} \quad (3)$$

Interpreting the results of the SI calculation is straightforward:

- SI > 0 indicates that mineral is supersaturated in solution and may precipitate onto aquifer matrix or pore space.
- SI $= 0$ indicates that mineral is at chemical equilibrium with the water.
- SI < 0 indicates that mineral is undersaturated in solution and may dissolve from aquifer matrix.

Due to potential systematic errors introduced during sampling and analysis, results within the range of ± 0.5 of zero are typically considered in or near chemical equilibrium.

4. Geochemical Model Input

To construct the equilibrium models in PHREEQC, site-specific data were used as input, including water chemistry, mineralogy, temperature, and pressure.

Data include the water chemistry data for the injection zones (Table 1) that were entered as received in ppm for elemental concentrations and standard units for pH.

In order to model the geochemistry of the clay minerals identified by XRD, an aluminum concentration was calculated in PHREEQC by equilibrating the provided water chemistry with the aluminosilicate clay mineral, smectite. The modeled aqueous concentration was used in subsequent modeling at 0.0275 ppm for CTV II. This concentration is reasonable for a sandstone aquifer at the neutral pH values.

For input into PHREEQC, the mineralogy in Table 2 was converted to a molar volume in moles per liter (mol/L) using porosity and bulk density values as follows:

- Injection Zone at CTV II, rock density of 2.65 kilograms per liter (kg/L) and porosity of 23 percent
- Upper Confining Zone at CTV II, rock density of 2.32 kg/L and porosity of 18.9 percent

The converted values for mineralogy that were input into PHREEQC are in shown Table 4.

Average temperature provided for the Injection Zone is 103°C at CTV II with an initial average pore volume pressure of 81.7 atm, which is expected to increase to 308.7 atm. The amount of carbon dioxide in 1 liter of gas at 176 atm and 100°C based on ideal gas law ($PV = nRT$) is 2.6 moles, and the amount of gas in 1 liter increases to 9.9 moles at 308.7 atm.

5. Geochemical Modeling Results and Discussion

Model results showing the changes in mineralogy designated as equilibrium phases in PHREEQC are presented for CTV II in Table 5 for the Injection Zone and in Table 6 for the Upper Confining Zone. Model results are presented in Table 7 for the water chemistry based on the equilibrium phases. The modeling steps were as follows:

- Injection Zone: Use the Injection Zone groundwater sample and equilibrate with zone mineralogy data set for the Injection Zone and carbon dioxide at given reservoir pressures and temperature. Models were run using each injectate composition.
- Upper Confining Zone: Use the model results for Injection Zone and equilibrate with the Upper Confining Zone mineralogy data set and carbon dioxide at final reservoir pressure. Models were run using each injectate composition.

Equilibrium geochemical modeling of the injection of carbon dioxide indicate that changes in mineralogy and aqueous chemistry are likely to occur, but overall, both geologic units are composed dominantly of silicate minerals such as quartz and feldspar that are not expected to be highly reactive during carbon dioxide sequestration. More reactive minerals like calcite and dolomite are present in relatively smaller amounts compared to the silicate minerals.

Although the model indicates that minerals will dissolve and precipitate, the net change in mass is minimal. Based on the molar mass, there is a small increase of less than 2 percent in the Injection Zone and a small increase of less than 1.5 percent in the Upper Confining Zone. These

changes indicate mineral precipitation is occurring during injection. The amount of porosity in the Injection Zone and Upper Confining Zone is not expected to be significantly impacted by mineral dissolution and precipitation reactions during carbon dioxide sequestration.

The TDS concentration is predicted to increase as dissolved aqueous species increase from the injection gases dissolving into the groundwater.

Based on the modeling, the following reactions are expected to occur:

- Dissolution of feldspars and calcite and the precipitation of quartz and siderite.
- Smectite and/or kaolinite dissolution, resulting in the precipitation of illite.
- Chlorite (chamosite) when initially present is not stable, and dissolves releasing iron, aluminum, and silica to solution.
- Anorthite when initially present is not stable, and dissolves releasing sodium, calcium, aluminum and silica to solution, likely contributing to calcite and clay mineral formation.
- Albite tends to be a stable feldspar mineral.
- Pyrite tends to dissolve releasing iron and sulfate to solution.

For both geologic units, the formation of carbonates like calcite, dolomite, or siderite was predicted to occur in several model scenarios. The formation of carbonate minerals can be an important mechanism to remove and immobilize carbon dioxide from solution through incorporation of CO₂ in the mineral phase.

The CO₂ gas in the injectate will form carbonate minerals, dissolve into solution, or remain in a gas phase.

Based on the equilibrium modeling, the aqueous chemistry results are provided in Table 7. Results indicate the following:

- Carbon dioxide will dissolve into solution and is included in the total inorganic carbon (TIC), which also includes bicarbonate and carbonate species. Results indicate that when carbon dioxide is dissolved in solution, the following dissolved species will occur as the following ions and complexes: carbon dioxide, bicarbonate, sodium bicarbonate, calcium bicarbonate, and magnesium bicarbonate.
- The pH values were around 5.5 in CTV II.

- The pe varied between oxidizing and reducing conditions for the various model runs.
- The calcium in solution includes the following ions and complexes: calcium, calcium bicarbonate, and calcium sulfate complex.

Based on the geochemical equilibrium modeling, the injection of carbon dioxide at the CTV II site does not cause significant reactions that will affect the injection or containment of the gas.

References

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Tables

Table 1. Baseline Geochemistry, CTV II Reservoir Formation

Analyte	Concentration (ppm ^a) at Produced Water Tank 1/19/2015
Barium	2
Bicarbonate	1,120
Calcium	118
Chloride	8,244
Magnesium	29.8
Potassium	85.3
pH (s.u.)	7.4
Silica	66
Sodium	5,967
Sulfate	4.9
Total dissolved solids	15,595

^a Unless otherwise noted

ppm = Parts per million

s.u. = Standard units

Table 2. Mineralogy for Upper Confining Zone, Injection Zone, and Lower Confining Zone

Zone	Depth (feet)	Mineralogical Content (%)													
		Quartz	Plagioclase	K-Feldspar	Calcite	Dolomite	Siderite	Barite	Pyrite	Kaolinite	Chlorite	Illite and Mica	Smectite	MXL I/S	Total Clay
Upper Confining Zone	8,828.0	23	21	9	3	—	—	—	1	12	5	—	—	26	43
	8,830.0	30	17	11	—	—	—	—	4	3	14	6	14		38
	8,909.0	20	20	13	—	—	—	—	2	5	3	—	—	35	43
	8,937.0	20	12	8	—	—	—	—	2	14	6	—	—	38	58
	8,939.0	24	18	11	1	—	—	—	3	3	15	8	17		43
	8,940.0	23	29	12	—	—	—	—	0	4	5	—	—	27	36
	8,942.0	23	15	10	—	—	—	—	2	12	5	—	—	33	50
	9,439.0	20	14	9	—	—	—	—	1	0	5	—	—	51	56
	9,441.0	21	19	12	2	—	—	—	3	0	0	—	—	43	43
Injection Zone	9,755.1	64	9	2	—	1	11	—	—	5	—	—	8	—	13
	9,758.5	70	12	4	—	1	2	—	—	5	—	—	6	—	11
	9,762.5	28	8	3	—	1	1	—	1	21	2	2	33	—	58
Lower Confining Zone	10,073.5	69	13	5	—	1	1	—	—	3	1	1	6	—	11
	10,077.5	30	7	2	1	—	3	—	1	17	5	2	32	—	56
	10,082.5	70	13	3	—	1	1	—		3	2	2	5	—	12
	10,090.5	51	8	2	—	1	—	—	2	8	4	3	21	—	36
	10,096.2	72	13	3	—	1	—	1	—	3	1	2	4	—	10
	10,070.5	69	14	4	—	1	1	—	—	4	1	—	6	—	11

— = Not detected

Table 3. Estimated Compositions for Carbon Dioxide Injectate

Gas	Mass Fraction (original composition)	Mass Fraction (normalized model input)
<i>Injectate Scenario 1</i>		
Carbon dioxide	0.9921253	0.99352
Nitrogen	0.0064308	0.00644
Hydrogen sulfide	0.0000295	0.00001
Sulfur dioxide	0.0000078	0.00003
Total	0.9985934	1.00
<i>Injectate Scenario 2</i>		
Carbon dioxide	0.9988419	0.9995
Methane	0.0003863	0.0004
Ethane	0.0005330	—
Hydrogen sulfide	0.0001394	0.0001
Total	0.9999007	1.00

Note: The original compositions were normalized to 100% for use as model input. For Scenario 2, the ethane component was excluded, as ethane is not in the model database.

Table 4. Mineralogy Input for PHREEQC Selected for Upper Confining Zone and Injection Zone

PHREEQC Mineral	Chemical Formula	Molar Mass (g/mol)	Input			
			Upper Confining Zone		Injection Zone	
			%	mol/L	%	mol/L
Quartz	SiO ₂	60.08	24	31.026	64	121.131
Albite (for plagioclase)	NaAlSi ₃ O ₈	263.02	18	5.315	9	3.891
K-Feldspar	KAlSi ₃ O ₈	278.33	11	3.070	2	0.817
Calcite	Ca(CO ₃)	100.09	1	0.776	0	0
Dolomite	CaMg(CO ₃) ₂	184.4	0	0	1	0.617
Siderite	Fe(CO ₃)	115.86	0	0	11	10.796
Pyrite	FeS ₂	119.98	3	1.942	0	0
Kaolinite	Al ₂ Si ₂ O ₅ (OH) ₄	258.16	3	0.906	5	2.202
Chamosite-7A	Fe ₂ Al ₂ SiO ₅ (OH) ₄	664.18	15.5	1.810	0	0
Illite	K _{0.6} Mg _{0.25} Al _{1.8} Al _{0.5} Si _{3.5} O ₁₀ (OH) ₂	389.34	7.7	1.544	0	0
Smectite-low-Fe-Mg	Ca _{.02} Na _{.15} K _{.2} Fe ⁺⁺ _{.29} Fe ⁺⁺⁺ _{.16} Mg _{.9} Al _{1.25} Si _{3.75} H ₂ O	549.07	16.8	2.372	8	1.657

Upper Confining Zone = 8,939 feet

Injection Zone = 9,755.1 feet

g/mol = Grams per mole

mol/L = Moles per liter

Table 5. Mineralogical Changes Based on Equilibrium Geochemical Modeling with Scenario 1 Injectate Chemistry

Mineral	Mineralogical Content (mol/L)								
	Initial	Final	Delta	Initial	Final	Delta	Initial	Final	Delta
<i>Pressure (atm)</i>	81.7			308.7			308.7		
<i>Sample</i>	<i>Injection Zone at 9755.1 feet</i>			<i>Injection Zone at 9755.1 feet</i>			<i>Confining Zone at 8939 feet</i>		
Albite	3.89	3.90	0.01	3.89	3.89	0.00	5.32	5.52	0.20
Anhydrite	—	—	—	—	—	—	0	0.50	0.50
CO ₂ (g)	2.62	2.67	0.06	9.89	10.22	0.33	9.89	3.84	-6.05
Calcite	0	0.34	0.34	0	0.34	0.34	0.78	0	-0.78
Chamosite-7A	0	0	0	0	0	0	1.81	0	-1.81
Dolomite	0.62	0.28	-0.34	0.62	0.28	-0.34	0	0.31	0.31
H ₂ S(g)	0.00003	0	-0.00003	0.00001	0	-0.00001	0.00001	3.37	3.37
Illite	0	1.36	1.36	0	1.37	1.37	1.54	5.53	3.99
K-Feldspar	0.82	0	-0.82	0.82	0	-0.82	3.07	0.96	-2.11
Kaolinite	2.2	1.05	-1.15	2.2	1.04	-1.16	0.91	0	-0.91
N ₂ (g)	0.0266	0.0263	-0.0003	0.1007	0.1007	-5.76 x 10 ⁻⁶	0.1007	0.1007	2.8 x 10 ⁻⁶
Pyrite	0	0	0	0	0	0	1.94	0	-1.94
Quartz	121.1	121.1	-0.006	121.1	121.1	0.006	31.03	31.9	0.87
SO ₂ (g)	0.00005	0	-0.0001	0.00002	0	-0.00002	0.00002	0	-0.00002
Siderite	10.8	10.8	0.001	10.8	10.8	0.0003	0	6.22	6.22
Smectite-low-Fe-Mg	1.66	1.66	-0.002	1.66	1.66	-0.0008	2.37	0.91	-1.46

Negative (-) delta value indicates that mineral or gas dissolves into solution, while positive (+) delta value indicates that mineral precipitates from solution.

mol/L = Moles per liter

atm = Atmospheres

— = Not an equilibrium phase

Table 6. Mineralogical Changes Based on Equilibrium Geochemical Modeling with Scenario 2 Injectate Chemistry

Mineral	Mineralogical Content (mol/L)								
	Initial	Final	Delta	Initial	Final	Delta	Initial	Final	Delta
<i>Pressure (atm)</i>	81.7			308.7			308.7		
<i>Sample</i>	<i>Injection Zone at 9755.1 feet</i>			<i>Injection Zone at 9755.1 feet</i>			<i>Confining Zone at 8939 feet</i>		
Albite	3.89	3.92	0.03	3.89	3.95	0.06	5.32	5.51	0.19
Anhydrite	—	—	—	—	—	—	0	0.48	0.48
CH ₄ (g)	0.003	0	0	0.01	0.00	-0.01	0.011	0	-0.011
CO ₂ (g)	2.64	2.49	-0.15	9.98	9.77	-0.21	9.98	3.90	-6.08
Calcite	0	0.21	0.21	0	0.00	0.00	0.78	0	-0.78
Chamosite-7A	0	0	0	0	0	0	1.81	0	-1.81
Dolomite	0.62	0.42	-0.20	0.62	0.63	0.01	0	0.33	0.33
H ₂ S(g)	0.0005	0	-0.0005	0.002	0.004	0.002	0.0018	3.38	3.38
Illite	0	1.41	1.41	0	1.52	1.52	1.54	5.55	4.01
K-Feldspar	0.82	0	-0.82	0.82	0.00	-0.82	3.07	0.95	-2.12
Kaolinite	2.2	1.08	-1.13	2.2	1.104	-1.096	0.91	0	-0.91
Pyrite	0	0	0	0	0	0	1.94	0	-1.94
Quartz	121.1	121.4	0.29	121.1	121.9	0.78	31.03	31.96	0.93
Siderite	10.80	10.87	0.07	10.8	11	0.1956	0	6.22	6.22
Smectite-low-Fe-Mg	1.66	1.50	-0.17	1.66	1.23	-0.435	2.37	0.89	-1.48

Negative (-) delta value indicates that mineral or gas dissolves into solution, while positive (+) delta value indicates that mineral precipitates from solution.

mol/L = Moles per liter

atm = Atmospheres

— = Not an equilibrium phase

Table 7. Modeled Equilibrium Aqueous Concentrations with Scenario 1 and Scenario 2 Injectates

Constituent	Concentration (mg/L ^a)					
<i>Injection Chemistry</i>	<i>Scenario 1</i>			<i>Scenario 2</i>		
<i>Geologic Zone</i>	<i>Injection Zone at 9755.1 feet</i>	<i>Confining Zone at 8939 feet</i>		<i>Injection Zone at 9755.1 feet</i>	<i>Confining Zone at 8939 feet</i>	
<i>Pressure (atm)</i>	81.7	308.7	308.7	81.7	308.7	308.7
Al ³⁺	0.208	0.211	0.190	0.208	0.211	0.189
Ba ²⁺	2.0	2.0	2.0	2.0	2.0	2.0
TIC	18,019	18,416	17,885	18,196	18,684	18,074
Ca ²⁺	76	62	9	77	12	9
Cl ⁻	8,373	8,373	8,373	8,373	8,373	8,373
Fe ²⁺	0.3	0.3	0.2	0.3	0.3	0.2
K ⁺	375	431	693	378	220	719
Mg ²⁺	14.76	12	65.10	15.12	48.91	77.99
N ₂	19	19	19	0	0	0
Na ⁺	5,821	5,844	6,357	5,837	5,883	6,481
SO ₄ ²⁻	1,202	1,204.6	2,589	1,242	1,059	2,924
SiO ₂	403.8	410.8	400.9	404.2	412.4	402.2
TDS (sum)	34,307	34,776	36,394	34,526	34,695	37,063
pH (s.u.)	5.5	5.5	5.6	5.5	5.5	5.6
pe (unitless)	1.7	1.5	-1.8	1.7	-1.9	-2.0

^a Unless otherwise noted
mg/L = Milligrams per liter
atm = Atmospheres
TDS = Total dissolved solids
s.u. = Standard units